Why does the interface of insulating oxides become conductive?

The ability to engineer electrical properties at semiconductor interfaces has facilitated the modern success of electronic technologies and is based on the concept of band alignment, which can be otherwise lifted in insulating oxides due to their characteristic wide band gaps and associated electronic properties [1]. The heterojunction between two insulating oxides is thus expected to remain insulating, unlike its semiconductor analog. However, the discovery of a metallic interface between insulating $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) in 2004 with large band gaps of 5.6 and 3.2 eV [2], respectively, has challenged this fundamental notion, thus launching the field of emergent phenomena at oxide interfaces. Determining the origin of this conductive interface has emerged as an unresolved problem in this context, and in 2016, our exploration of head-to-head ferroelectric-like polarizations across the metallic interface shed new light on this long-standing puzzle [1].

Indeed, continuous efforts have been devoted to disentangling the problem using state-of-the-art characterizationtechniques, ranging from synchrotron-based spectroscopies and surface X-ray diffraction to electron microscopy and spectroscopy at a high spatial resolution [1]. Several scenarios have been readily proposed, including electrostatic-driven interfacial charge transfer (popularly known as polar catastrophe), a chemical-intermixing-induced metallic phase at the interface, electron doping due to oxygen vacancies in STO, and structural modulations in interfacial lattices [1]. Nevertheless, none of these proposed models was found to be robust [1], and the underlying reason could be the lack of quantitative accuracy with atomic-level precision in all these characterization techniques. To determine why the LAO/STO interface can become conductive, it is crucial to quantitatively examine the chemical, electronic, and structural properties of all individual interfacial lattices with the highest possible atomic accuracy.

Using atomic-scales canning transmission electron microscopy (STEM) and electron energy-loss spectroscopy (EELS) established at National Taiwan University, which represents the only functional setup of this kind in Taiwan, we simultaneously quantified the chemical composition of individual unit cells, estimated the valence of transition-metal atoms, and precisely evaluated the picometer-scale structural distortions across interfacial lattices. Achieving all these measurements simultaneously is unparalleled by any other existing methods [1]. Quantitatively scrutinizing all experimental results on LAO/STO at atomic-level accuracy (e.g., Figure 1a) then indicated that the previous proposals of polar catastrophe, intermixing, oxygen vacancies, and structural modulations were rather irrelevant to the interfacial conductivity. Instead, the metallic interface is consistently accompanied by ferroelectric-like polarizations across the interface (Figure 1b), whereas such a lattice instability is forbidden inthe primitive LAO and STO. A thorough exploration of the corresponding energetic landscapes determined that ferroelectric-like lattice instabilities can indeed be rejuvenated by the experimentally observed strained condition in the interfacial unit cells (e.g.,Figure 1a). Most importantly, the lattice instabilities form a head-to-head configuration (Figure 1b) with divergent depolarization fields, rendering the interface an electron reservoir. Electronic carriers readily accumulate at the interface, forming a metallic two-dimensional electron gas (2DEG) while assisting in the screening of the polar divergence.

Notably, the emergence of this distinct conductive LAO/STO interface arises from the unprecedented scope of strain-rejuvenated head-to-head polar lattice instabilities, and our electronic and structural quantifications at an atomic precision played a crucial role in the discovery. This comprehensive work, which was completely accomplished in Taiwan, is anticipated to stimulate the design of oxide heterostructures from novel structural aspects in the future.

References

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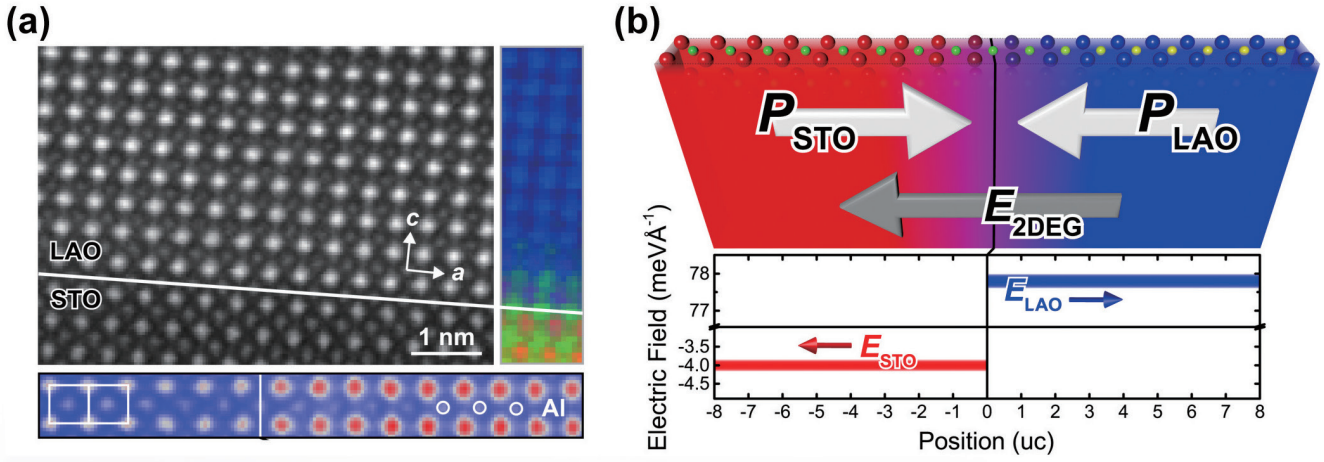

Figure 1. (a) STEM imaging of the conductive LAO/STO interface (top left) and corresponding STEM-EELS chemical mapping (top right; green, Ti; red, Sr; blue, La) at atomic resolution. Bottom panel, magnified STEM image with the *c*-axis pointing along the horizontal direction and revealing the tetragonal distortion of STO and ferroelectric-like off-center distortion of Al in LAO. (b) The rejuvenation of hidden ferroelectric-like lattice instabilities, P_{LAO} and P_{STO}, across the LAO/STO interface due to the characteristic epitaxial strain with the head-to-head polar divergence causing the interface to become an electron reservoir, thereby readily harboring the screening of 2DEG. E_{2DEG} is the accompanying electric field.